Development of hybrid L-edge/XRF densitometer for nuclear material solution assay

Seunghoon Park [1*], Sung-Woo Kwak[1], Jerry Park[2], Uk-Rayng Park[1], Ae-Ri Lee[1], and Heejun Chung[1]
[1]*Korea Institute of Nuclear Nonproliferation and Control, Yuseoungdaero 1534, Yuseong-gu, Daejeon, Repulic of Korea*
[2] *ISP Co., Ltd, 422 Palbok-dong 3-ga, Deokjin-gu, Jeonju-si, Jeollabuk-do, Republic of Korea*
[*]*Corresponding author: shpark@kinac.re.kr*

1. Introduction

THE low energy X-ray measurement system is used for on-site analytical measurements of safeguards in various nuclear facilities, such as nuclear power plant and reprocessing plants. Specially, the hybrid system of X-ray absorption spectrometry such as L-edge densitometry (LED) [1, 2], and X-ray fluorescence spectrometry (XRF) has an important role of safeguards for nuclear facilities. LED is a technique of determination of uranium concentration as a continuous X-ray energy beams transmit a uranium liquid sample for safeguards. Compared to K-edge densitometer [3], due to relatively lower energy (L-edge energy is 17.17 keV) of Uranium L-series energy than K-series energy, L-edge densitometer does not require high purity germanium detector with liquid nitrogen cooling. Therefore, the L-edge densitometer is appropriate for portable equipment for on-site nuclear material inspection and safeguards at facility sites. XRF combined with LED is a technique of finding of nuclear materials from reflected characteristic X-ray photons. In this study, the hybrid system of L-edge and XRF system (HLED) was developed for determination of concentration of nuclear materials. We verified that the system has feasibility of determination of the concentration of non-radio nuclide from analysis of a surrogate material such as led (Pb) solutions prior to the work of nuclear materials.

2. Methods and Results

2.1 Hybrid L-edge/XRF densitometer

The hybrid LED/XRF densitometer (HLED) consists of an X-ray tube, shields, a sample container, and detectors. The detailed design is shown in Fig. 1. The detectors are set up at front and normal of a sample container for L-edge densitometry. Both of detectors are silicon drift detectors (SDD) which 25mm^2 × 500 μm. The diameter of X-ray photon starting point to the sample is 1.6 mm in front of collimator with hole of 3 mm-diameter and length of 5-cm-long. The sample container consist of the 45 ml volume component for XRF to maximize detection efficiency connected with small volume component of 2 mm-optical path length shown in Fig. 3. There are two beam lines from the X-ray tube to each sample volume. The X-ray tube can be operated by 35 kVp and 100 μA with rhodium (Rh) anode.

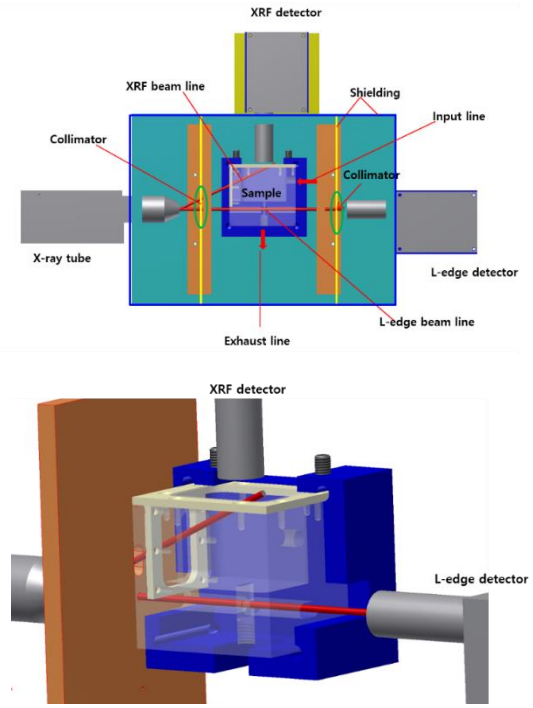

Fig. 1 Design of HLED and two beam line for L-edge densitometry and X-ray fluorescence spectrometry

X-ray photons from the tube transmit across or randomly react the sample with characteristic energy after interaction with the sample material. The transmission and emitted fluorescence photon energy spectra are recorded by the silicon drift detector.

The system was fabricated based on previous study [4] of Monte Carlo simulation for the instrument design and feasibility study of determination of nuclear material concentration. The prototype of HLED system is shown in Fig. 2

Fig. 2 The prototype of hybrid L-edge/XRF (HLED) system.

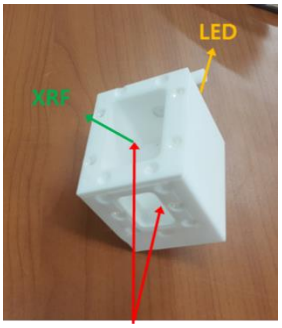

Fig. 3 The sample container for L-edge and XRF analysis

2.2 Analysis of L-edge of surrogate material

Fig. 4 is the simulated energy spectrum of x-ray transmitted nitric acid blanket solution and the led nitric acid solution. The material (led - Pb) concentration is assumed that led solid samples into a nitric acid solution. The jump of energy spectrum transmitted through led solution is observed compared to reference energy spectrum transmitting across liquid nitric acid. The height of the jump of the spectrum determines the sample concentrations. The first jump of the spectrum is 13.06 keV, L_{III} energy (L-edge). Because the height is larger than other two energy jumps, L_{III} jump is used to analysis of led concentration

Material concentrations through transmitted X-ray can be calculated following equation.

$$\rho_{Pb} = \ln\left[\frac{T(E_-)/T(E_+)}{\Delta\mu D}\right]$$

$T(E_-)$, $T(E_+)$ are respectively transmission at the energies E_- (Lower than L_{III}) and E_+ (Upper than L_{III}). D is sample thickness (exactly, optical path length) and $\Delta\mu$ is mass attenuation coefficient difference at each transmission energy. Extrapolated fitting in linearized a representation lnln(1/T) vs ln E is applied for determining upper and lower transmission for L-edge. Fitting intervals range from 11 – 12 keV for E_- and 14 – 15 keV for E_+. The fitting is shown in Fig. 5.

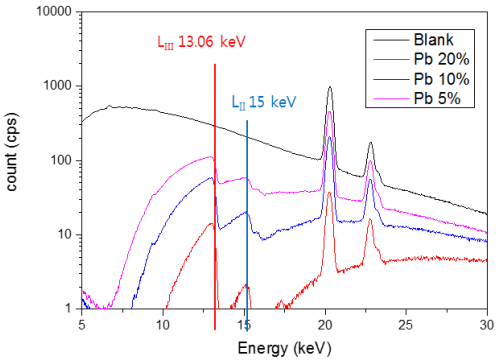

Fig. 4 Transmitted spectrum through led solution for led concentration variation

The height of jump of the spectrum is determined by the uranium concentration shown in Fig. 5. The height of jump is dependent of uranium density. The spectra with high concentrated material has large height of jump.

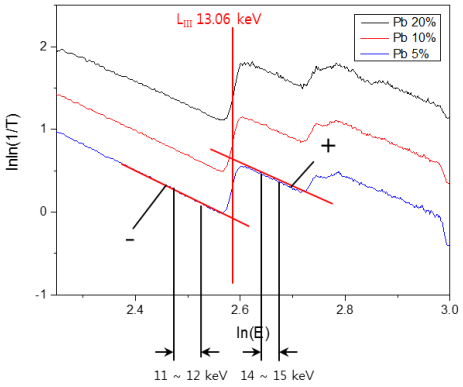

Fig. 5 Extrapolated fitting curve in lnln(1/T) vs ln E

The estimated concentration from the experiments is shown in table I. The errors, < 2%, are rather higher than ITV 0.7%. Due to the led solutions can be incorrect due to no precise measurements except for L-edge densitometry. The standard samples are required in order to enhance accuracy of the L-edge analysis.

TABLE I. ESTIMATED LED CONCENTRATION FROM EXPERIMENT

Declared (g/cm^3)	Estimation (g/cm^3)
0.050	0.055
0.100	0.102
0.200	0.204

3. Conclusions

From this study, the proto type of a hybrid L-edge/XRF densitometer (HLED) was developed for nuclear material solution assay. L-edge analysis shows feasibility of the system by analysis of non-radioactive surrogate material (Pb) solution. Based on the study, nuclear material will be analyzed and reduce uncertainty by analysis of non-radioactive and radioactive standard samples such as led and uranium nitric acid solutions.

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